REMARKS

Applicants thank the Examiner for the thorough examination given the present application.

Status of the Claims

Claims 1-10 and 12-16 are pending in the above-identified application. Claims 1 and 10 have been amended. Support for the recitations in claims 1 and 10 can be found in the publication of the present specification, *inter alia*, at paragraph [0026]. Thus, no new matter has been added.

Applicants submit that the present Amendment reduces the number of issues under consideration and places the case in condition for allowance. Alternatively, entry of the present amendment is proper to place the claims in better form for appeal.

In view of the following remarks, Applicants respectfully request that the Examiner withdraw all rejections and allow the currently pending claims.

Drawings

Applicants thank the Examiner for indicating that the drawings are accepted.

Issues under 35 U.S.C. § 103(a)

1) Claims 1-6 and 8-9 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Mizumoto et al. '263 (US 4,631,263).

2) Claim 7 is rejected under 35 U.S.C. § 103(a) as being unpatentable over Mizumoto et al. '263 in view of Nishino et al. '355 (JP 55-149355).

3) Claims 10 and 12-16 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Yokota et al. '063 (US 4,625,063) in view of Mizumoto et al. '263.

Applicants respectfully traverse. Reconsideration and withdrawal of the rejections are respectfully requested based on the following considerations.

Legal Standard for Determining Prima Facie Obviousness

MPEP 2141 sets forth the guidelines in determining obviousness. First, the Examiner has to take into account the factual inquiries set forth in *Graham v. John Deere*, 383 U.S. 1, 17, 148 USPQ 459, 467 (1966), which has provided the controlling framework for an obviousness analysis. The four *Graham* factors are:

(a) determining the scope and content of the prior art;

(b) ascertaining the differences between the prior art and the claims in issue;

(c) resolving the level of ordinary skill in the pertinent art; and

(d) evaluating any evidence of secondary considerations.

Graham v. John Deere, 383 U.S. 1, 17, 148 USPQ 459, 467 (1966).

Second, the Examiner has to provide some rationale for determining obviousness. MPEP 2143 sets forth some rationales that were established in the recent decision of *KSR International Co. v Teleflex Inc.*, 82 USPQ2d 1385 (U.S. 2007). Exemplary rationales that may support a conclusion of obviousness include:

(a) combining prior art elements according to known methods to yield predictable results;

(b) simple substitution of one known element for another to obtain predictable results;

(c) use of known technique to improve similar devices (methods, or products) in the same way;

(d) applying a known technique to a known device (method, or product) ready for improvement to yield predictable results;

(e) "obvious to try" – choosing from a finite number of identified, predictable solutions, with a reasonable expectation of success

(f) known work in one field of endeavor may prompt variations of it for use in either the same field or a different one based on design incentives or other market forces if the variations are predictable to one of ordinary skill in the art;

(g) some teaching, suggestion, or motivation in the prior art that would have led one of ordinary skill to modify the prior art reference or to combine prior art reference teachings to arrive at the claimed invention.

As the MPEP directs, all claim limitations must be considered in view of the cited prior art in order to establish a *prima facie* case of obviousness. *See* MPEP 2143.03.

Distinctions over the Cited References

In the outstanding Office Action, the Examiner acknowledges the previous argument that Mizumoto et al. '263 fail to disclose that the interior of the catalyst can be used as sites for the reaction. However, the Examiner asserts that this feature is not recited in the claims. As amended, claim 1 now recites that "the inside of the catalyst is a site of reaction for producing said tertiary amine," and claim 10 now recites "a step of reacting an alcohol with a primary or secondary amine inside a film-type catalyst." The cited references fail to disclose these features.

On page 4 of the outstanding Office Action, the Examiner states, "Such water-repellent catalysts are used to wide range of gas/liquid reactions. Gas/liquid reactions include reactants consisting of gas and liquid and effect a chemical conversion between these reactants (col.2, lines 50-58). It meets the instant claimed limitation of the mass (i.e. gas) transfer between the inside and outside of the catalyst.

In the present specification, the reactants of the amination reaction may be present in a gaseous or liquid phase (paragraph [0060] of the present published application). The process of transferring the reactants and the product in the inside of the catalyst is governed by diffusion, and the distance is reduced to 500 μm or less, whereby the mass transfer between the inside and outside of the catalyst can be promoted thereby effectively utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of the intermediate reaction product in the inside of the catalyst (paragraph [0026] of the present published application). The diffusivity, which governs the mass transfer of reactants and product in the inside of the catalyst, has been well acknowledged to be much smaller in the liquid state than in the gaseous state, as disclosed in many articles. For example, in the enclosed **Bird et al.** reference (*Transport Phenomena*), some typical diffusivity coefficients in a gaseous state are disclosed to be of the order of 0.1 cm^2/sec (see page 503, **Table 16.2-2**), while those in a liquid state are of the order of 10^5 cm^2/sec (see page 504, **Table 16.2-3**). As such, the film type catalyst of the present invention is much more effective for producing tertiary amine than Mizumoto et al. '263, especially when the reactants and product are in a liquid state.

Moreover, claims 1 and 10 specifically recite that "said film-type catalyst comprises catalyst particles bound to one another via a synthetic resin as a binder" and that "said particles form a three-dimensional network structure via the binder on a substrate." Accordingly, Mizumoto et al. '263 fail to disclose the substrate as recited in the pending claims.

Furthermore, claims 1 and 10 recite "whereby the diffusion rate in the catalyst layer is increased due to said three-dimensional network structure, and the mass transfer between the inside and outside of the catalyst can be promoted thereby utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of an intermediate reaction product in the inside of the catalyst."

The Examiner relies on column 2, lines 27-32 of Mizumoto et al. '263, which states, "When the water-repellent catalyst of this invention is used, it becomes possible that the gas passes not only over the surfaces of the catalyst but also through the interior of the catalyst and, accordingly, three-phase interfaces are easily formed, and the rate of reaction can be increased." As noted above, the present invention utilizes the whole of the catalyst. In contrast, Mizumoto et al. '263 only uses the interior of the catalyst for permeating the gas. Mizumoto et al. '263 fail to disclose that the interior of the catalyst can be used as sites for the reaction. In fact, Mizumoto et al. '263 state that the liquid does not penetrate into the catalyst because of liquid impermeability (col. 2, lines 18-19).

The gas/liquid reactions in the presence of a water-repellent catalyst proceed through the formation of three-phase interfaces on the surface of the catalyst, and the inside of the catalyst is never or scarcely used as sites for the reaction (col. 2, lines 34-38). The gas/liquid reactions that proceed in the presence of a water-repellent catalyst, such as an isotopic exchange reaction between water and hydrogen gas, require a gas and a liquid component to contact with the solid catalyst. Water cannot exchange its hydrogen atoms with hydrogen gas unless it is in contact with the solid catalyst. A water molecule must trap its hydrogen atom at a reaction site on the surface of the catalyst to get a substitute one trapped at another site in order to exchange them. The catalyst assembly of Mizumoto et al. '263 can be effectively utilized by letting the gas permeate the porous interior to reach the surface and then the reaction proceeds through the formation of three-phase interfaces.

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In contrast, the present invention is directed to permeability of reactants and products through the porous catalyst layer. Then, the reaction proceeds as evidenced by the comparison of Examples 4 and 5, generating more by-products with a thicker catalyst (see Table 2 on page 43 of the present specification). The reason why a thicker catalyst layer produces worse selectivity of the tertiary amine is explained in the present specification. The present specification recites that "the process of transferring the reactants and the product in the inside of the catalyst is governed by diffusion, and the distance is reduced to 500 μm or less, whereby the mass transfer between the inside and outside of the catalyst can be promoted thereby effectively utilizing the whole of the catalyst and simultaneously suppressing the excessive reaction of the intermediate reaction product in the inside of the catalyst" (paragraph [0026] of the publication of the present application). If the reaction proceeded only on the surface of the film-type catalyst as on the surface of the catalyst assembly of Mizumoto et al. '263, the thickness of the film-type catalyst would not influence the reactivity.

This phenomenon is illustrated by comparing Example 1 and Comparative Example 1 (see Table 1 on page 42 of the present specification). The difference in the way the catalysts are prepared (which creates a difference in their internal structures) and the difference in thickness of the catalysts lead to very different results for the amination reaction. The film-type catalyst of Example 1 generates 95% of the desired amine with only 4% of undesired amine in five hours. In contrast, the pellet-type catalyst of Comparative Example 1, which has almost thirty times as much in weight as the film-type catalyst of Example 1, generates only 60% of the desired amine with 5% of the undesired amine. **As such, the film-type catalyst of the present invention can utilize the whole of the catalyst and simultaneously suppress the excessive reaction of the intermediate reaction product in the inside of the catalyst. This phenomenon is totally different from Mizumoto et al. '263 as described above.**

Turning to claims 3 and 13, the Examiner asserts that Mizumoto et al. '263 disclose a catalyst comprising copper. Applicants respectfully traverse. Mizumoto et al. '263 only disclose Pt, Ru, Rh, Ir, or an alloy thereof as a catalytically active noble metal (col. 3, lines 33-35).

Turning to claims 8 and 15, the Examiner relies on Mizumoto et al. '263 to disclose the subject matter of this claim. Applicants respectfully traverse. The substrate of Mizumoto et al. '263 is not a metal film. Rather, the substrate is a water-repellent porous polytetrafluoroethylene.

As discussed above, Mizumoto et al. '263 do not disclose each and every aspect of claims 1 and 10, from which all other claims ultimately depend. Applicants respectfully submit that Nishino et al. '355 and Yokota et al. '063 do not overcome the deficiencies of this reference.

To establish a *prima facie* case of obviousness of a claimed invention, all of the claim limitations must be disclosed by the cited references. As discussed above, the cited references fail to disclose all of the claim limitations of independent claims 1 and 10, and those claims dependent thereon. Accordingly, the combination of references does not render the present invention obvious.

Furthermore, the cited references or the knowledge in the art provide no reason or rationale that would allow one of ordinary skill in the art to arrive at the present invention as claimed. Therefore, a *prima facie* case of obviousness has not been established, and withdrawal of the outstanding rejections is respectfully requested. Any contentions of the USPTO to the contrary must be reconsidered at present.

Conclusion

All of the stated grounds of rejection have been properly traversed, accommodated, or rendered moot. Applicants therefore respectfully request that the Examiner reconsider all presently outstanding rejections and that they be withdrawn. It is believed that a full and complete response has been made to the outstanding Office Action, and as such, the present application is in condition for allowance.

Should there be any outstanding matters that need to be resolved in the present application, the Examiner is respectfully requested to contact Chad M. Rink, Registration No. 58,258, at the telephone number of the undersigned below to conduct an interview in an effort to expedite prosecution in connection with the present application.

If necessary, the Director is hereby authorized in this, concurrent, and future replies to charge any fees required during the pendency of the above-identified application or credit any overpayment to Deposit Account No. 02-2448.

Dated: November 19, 2010

Respectfully submitted,

By
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Exhibit I: R. B. Bird, et al., *Transport Phenomena*, John Wiley & Sons, Inc., New York, 1960 (pages 502-505).

Wiley International Edition

TRANSPORT PHENOMENA

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Preface

This book is intended to be an introduction to the field of transport phenomena for students of engineering and applied science. Herein we present the subjects of momentum transport (viscous flow), energy transport (heat conduction, convection, and radiation), and mass transport (diffusion). In this treatment the media in which the transport phenomena are occurring are regarded as continua, and very little is said about the molecular explanation of these processes. Surely the continuum approach is of more immediate interest to engineering students, although it should be emphasized that both approaches are needed for complete mastery of the subject.

Because of the current demand in engineering education to put more emphasis on understanding basic physical principles than on the blind use of empiricism, we feel there is a very definite need for a book of this kind. Obviously the subject matter is sufficiently basic that it cuts across traditional departmental lines. Our thought has been that the subject of transport phenomena should rank along with thermodynamics, mechanics, and electromagnetism as one of the key "engineering sciences." Knowledge of the basic laws of mass, momentum, and energy transport has certainly become important, if not indispensable, in engineering analysis. In addition, the material in this text may be of interest to some who are working in physical chemistry, soil physics, meteorology, and biology.

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§16.2 FICK'S LAW OF DIFFUSION

In Eq. 1.1–2 the viscosity μ is defined as the proportionality factor between momentum flux and velocity gradient (Newton's law of viscosity). In Eq. 8.1–6 the thermal conductivity k is defined as the proportionality factor between heat flux and temperature gradient (Fourier's law of heat conduction).

TABLE 16.2–1
EQUIVALENT FORMS OF FICK'S FIRST LAW OF BINARY DIFFUSION

Flux	Gradient	Form of Fick's First Law	
n_A	$\nabla \omega_A$	$n_A - \omega_A(n_A + n_B) = -\rho \mathscr{D}_{AB} \nabla \omega_A$	(A)
N_A	∇x_A	$N_A - x_A(N_A + N_B) = -c \mathscr{D}_{AB} \nabla x_A$	(B)
j_A	$\nabla \omega_A$	$j_A = -\rho \mathscr{D}_{AB} \nabla \omega_A$	(C)
$J_A^\star$	∇x_A	$J_A^\star = -c \mathscr{D}_{AB} \nabla x_A$	(D)
j_A	∇x_A	$j_A = -\left(\dfrac{c^2}{\rho}\right) M_A M_B \mathscr{D}_{AB} \nabla x_A$	(E)
$J_A^\star$	$\nabla \omega_A$	$J_A^\star = -\left(\dfrac{\rho^2}{cM_A M_B}\right) \mathscr{D}_{AB} \nabla \omega_A$	(F)
$c(v_A - v_B)$	∇x_A	$c(v_A - v_B) = -\dfrac{c \mathscr{D}_{AB}}{x_A x_B} \nabla x_A$	(G)

Now we define the mass diffusivity $\mathscr{D}_{AB} = \mathscr{D}_{BA}$ in a binary system in an analogous fashion:

$$J_A^\star = -c\mathscr{D}_{AB}\nabla x_A \qquad (16.2\text{-}1)$$

This is Fick's first law of diffusion,[1] written in terms of the molar diffusion flux $J_A^\star$. This equation states that species A diffuses (moves relative to the mixture) in the direction of decreasing mole fraction of A, just as heat flows by conduction in the direction of decreasing temperature.

A number of other mathematically equivalent statements of Fick's first law have appeared in the literature, and some of them are summarized in Table 16.2–1 for reference only. The diffusivity $\mathscr{D}_{AB}$ is identical in all these equations. Of special importance in the following chapters is the form of Fick's first law in terms of N_A, the molar flux relative to stationary coordinates:

$$N_A = x_A(N_A + N_B) - c\mathscr{D}_{AB}\nabla x_A \qquad (16.2\text{-}2)$$

This equation shows that the diffusion flux N_A relative to stationary coordinates is the resultant of two vector quantities: the vector $x_A(N_A + N_B)$,

[1] Temperature gradients, pressure gradients, and external forces also contribute to the diffusion flux, although their effects are usually minor. More complete expressions for the diffusion flux are given in §18.4.

Fick's Law of Diffusion

which is the molar flux of A resulting from the bulk motion of the fluid, and the vector $J_A^\star = -c\mathscr{D}_{AB}\nabla x_A$, which is the molar flux of A resulting from the diffusion superimposed on the bulk flow. Thus in Fig. 16.1–1b the bulk flow and diffusion terms in Eq. 16.2–2 are in the same direction for species A (because A is diffusing with the current) and are opposed for species B (because B is diffusing against the current).

The units of the mass diffusivity $\mathscr{D}_{AB}$ are $cm^2\,sec^{-1}$ or $ft^2\,hr^{-1}$. Note that the kinematic viscosity ν and the thermal diffusivity α also have the same units. The way in which these three quantities are analogous can be seen from the following equations for the fluxes of mass, momentum, and energy in one-dimensional systems:

$$j_{Ay} = -\mathscr{D}_{AB}\frac{d}{dy}(\rho_A) \qquad \text{(Fick's law}^1 \text{ for constant } \rho) \qquad (16.2\text{-}3)$$

$$\tau_{yx} = -\nu\frac{d}{dy}(\rho v_x) \qquad \text{(Newton's law for constant } \rho) \qquad (16.2\text{-}4)$$

$$q_y = -\alpha\frac{d}{dy}(\rho \hat{C}_p T) \qquad \text{(Fourier's law for constant } \rho\hat{C}_p) \qquad (16.2\text{-}5)$$

These equations state, respectively, that (a) mass transport occurs because of a gradient in mass concentration, (b) momentum transport occurs because of a gradient in momentum concentration, and (c) energy transport occurs because of a gradient in energy concentration. These analogies do not apply in two- and three-dimensional problems, however, because τ is a tensor quantity with nine components, whereas j_A and q are vectors with three components.

In Tables 16.2–2, 3, and 4 some values of $\mathscr{D}_{AB}$ are given for a few gas,

TABLE 16.2–2
EXPERIMENTAL DIFFUSIVITIES OF SOME DILUTE GAS PAIRS[a]

Gas Pair	Temperature (°K)	$\mathscr{D}_{AB}$ (cm² sec⁻¹)
CO₂–N₂O	273.2	0.096
CO₂–CO	273.2	0.139
CO₂–N₂	273.2	0.144
	288.2	0.158
	298.2	0.165
Ar–O₂	293.2	0.20
H₂–SF₆	298.2	0.420
H₂–CH₄	298.2	0.726

[a] This table is abstracted from J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, Molecular Theory of Gases and Liquids, Wiley, New York (1954), p. 579. The values given are for 1 atm pressure.

TABLE 16.2–3

EXPERIMENTAL DIFFUSIVITIES IN THE LIQUID STATE[a]

A	B	$T(^\circ C)$	x_A	$\mathscr{D}_{AB} \times 10^5$ $(cm^2\ sec^{-1})$
Chlorobenzene	Bromobenzene	10.01	0.0332	1.007
			0.2642	1.069
			0.5122	1.146
			0.7617	1.226
			0.9652	1.291
		39.97	0.0332	1.584
			0.2642	1.691
			0.5122	1.806
			0.7617	1.902
			0.9652	1.996
Ethanol	Water	25	0.05	1.13
			0.275	0.41
			0.50	0.90
			0.70	1.40
			0.95	2.20
Water	n-Butanol	30	0.131	1.24
			0.222	0.920
			0.358	0.560
			0.454	0.437
			0.524	0.267

[a] This table is abstracted from a review article by P. A. Johnson and A. L. Babb, Chem. Revs., 56, 387–453 (1956); in this article a summary of experimental diffusion coefficients for liquid systems is given, as well as a survey of methods of measurement. Another excellent review article is that of L. J. Gosting, Measurement and Interpretation of Diffusion Coefficients of Proteins, Advances in Protein Chemistry, Vol. XI, Academic Press, New York (1956).

liquid, and solid systems. Diffusivities of gases at low density are almost composition independent, increase with the temperature, and vary inversely with pressure. Liquid and solid diffusivities are strongly concentration-dependent and generally increase with temperature. In the next three sections we summarize the available means for estimating diffusivities.

§16.3 TEMPERATURE AND PRESSURE DEPENDENCE OF MASS DIFFUSIVITY

The mass diffusivity $\mathscr{D}_{AB}$ for a binary system is a function of temperature, pressure, and composition, whereas the viscosity μ and thermal conductivity

TABLE 16.2–4

EXPERIMENTAL DIFFUSIVITIES IN THE SOLID STATE[a]

System	$T(^\circ C)$	Diffusivity, $\mathscr{D}_{AB}$ $(cm^2\ sec^{-1})$
He in SiO_2	20	$2.4 - 5.5 \times 10^{-10}$
He in pyrex	20	4.5×10^{-11}
	500	2×10^{-8}
H_2 in SiO_2	500	$0.6 - 2.1 \times 10^{-8}$
H_2 in Ni	85	1.16×10^{-8}
	165	10.5×10^{-8}
Bi in Pb	20	1.1×10^{-16}
Hg in Pb	20	2.5×10^{-15}
Sb in Ag	20	3.5×10^{-21}
Al in Cu	20	1.3×10^{-30}
Cd in Cu	20	2.7×10^{-15}

[a] Values taken from R. M. Barrer, Diffusion in and through Solids, Macmillan, New York (1941), pp. 141, 222, and 275.

k for a pure fluid are functions only of temperature and pressure. The data available on $\mathscr{D}_{AB}$ for most binary mixtures are, moreover, quite limited in range and accuracy. The available correlations of $\mathscr{D}_{AB}$ are of limited scope and are based more on theory than on experiment.

For binary gas mixtures at low pressure, $\mathscr{D}_{AB}$ is inversely proportional to the pressure, increases with increasing temperature, and is almost independent of composition for a given gas-pair. The following equation for estimation of $\mathscr{D}_{AB}$ at low pressures has been developed[1] from a combination of kinetic theory and corresponding-states arguments:

$$\frac{p\mathscr{D}_{AB}}{(p_{cA}p_{cB})^{1/3}(T_{cA}T_{cB})^{5/12}\left(\frac{1}{M_A}+\frac{1}{M_B}\right)^{1/2}} = a\left(\frac{T}{\sqrt{T_{cA}T_{cB}}}\right)^b \qquad (16.3\text{–}1)$$

in which $\mathscr{D}_{AB}$ [=] cm² sec⁻¹, p [=] atm, and T [=] °K. Analysis of experimental data gave the following values of the constants a and b:

For nonpolar gas-pairs:

$$a = 2.745 \times 10^{-4}$$
$$b = 1.823$$

For H_2O with a nonpolar gas:

$$a = 3.640 \times 10^{-4}$$
$$b = 2.334$$

[1] J. C. Slattery and R. B. Bird, A.I.Ch.E. Journal, 4, 137–142 (1958).